



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of Syuichi Ikenoue et al.

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Examiner: Kishor Mayekar

For: Multi-Layer Coating Film-Forming Method

DECLARATION UNDER RULE 37 C.F.R. 1.132

HONORABLE COMMISSIONER OF PATENTS AND TRADEMARKS

WASHINGTON, D.C. 20231

Sir:

Now comes Syuichi Ikenoue, who deposes and says that:

1. I am one of the inventors of the present invention of this patent application.

2. The following experiments shown by Experiment 1 and Comparative Experiments 1-3 were carried out by me or my direct supervision and control.

3. Experiments were carried out as follows:

Preparation Experiment 1

Preparation of Cationic Electrodeposition Coating Composition No. 1 (corresponding to Cationic Electrodeposition Coating Composition (A-2) in the present invention):

Into 450 parts of butyl cellosolve was dissolved 1260 parts of bisphenol A based epoxy resin having an epoxy equivalent of 630 (Epikote 1002, trade name, marketed by

Shell Kagaku K.K.), followed by adding 132 parts of p-nonyl phenol and 105 parts of N-methyl ethanolamine, heating up to 140°C, and reacting at that temperature to obtain an addition epoxy resin having a solid content of 77% and an amine value of 52. To 130 parts of the resulting resin were added 30 parts of blocked polyisocyanate compound (as a curing agent) and 1.3 parts of polypropylene glycol (number average molecular weight 4000), followed by adding 2.1 parts of acetic acid to make water-soluble, slowly adding deionized water, and dispersing to obtain an emulsion having a solid content of 30%.

On the other hand, 4.7 parts of 75% epoxy based amine type pigment-dispersing resin was neutralized with 0.16 part of 88% aqueous formic acid solution, followed by adding 22.2 parts of deionized water, adding 15 parts of titanium white, 7 parts of clay, 0.3 part of carbon black, 3.0 parts of bismuth hydroxide (2.4 parts as bismuth metal) and 3 parts of dioctyltin oxide, dispersing in a ball mill to obtain a pigment dispersed paste having a solid content of 55%.

Thereafter, the emulsion having the solid content of 30% was mixed with the pigment dispersed paste having the solid content of 55%, followed by diluting with deionized water so as to obtain a cationic electrodeposition coating composition No. 1 having a solid content of 19%.

The above blocked polyisocyanate was prepared by reacting a reaction product between 174 parts of 2,6-tolylene diisocyanate and 85 parts of polycaprolactone diol having a

hydroxy equivalent of 425 with 2-ethylhexyl alcohol monoether (blocking agent) of ethylene glycol.

#### Preparation Experiment 2

Preparation Experiment 1 was duplicated except that 3.4 parts (2.4 parts as bismuth metal) of bismuth lactate was used in place of 3 parts of bismuth hydroxide to obtain an electrodeposition coating composition No. 2.

#### Preparation Experiment 3

Preparation Experiment 1 was duplicated except that 2.7 parts (2.4 parts as bismuth metal) of  $\text{Bi}_2\text{O}_3$  was used in place of 3 parts of bismuth hydroxide to obtain an electrodeposition coating composition No. 3.

#### Preparation Experiment 4

Preparation of Water Based Intercoat Coating Composition No. 1 (corresponding to Water Based Intercoat Coating Composition ( $\text{B}_1\text{-i}$ ) in the present invention):

A mixture of 70 parts of polyester resin (Note 1), 4 parts of dimethyl aminoethanol, 30 parts of aliphatic hexafunctional blocked polyisocyanate compound (Note 2), 10 parts of fine aluminum powder coated with a phosphate group-containing compound (Note 3), 60 parts of titanium oxide white pigment (Note 5) and one part of carbon black was dispersed into deionized water to obtain a water based intercoat coating composition ( $\text{B}_1\text{-i}$ ) so as to have a viscosity of 10 seconds (Ford-cup #4/20°C) and a solid content of 15%.

(Note 1) Polyester resin: A reactor was charged with 756

parts of neopentyl glycol, 109 parts of trimethylolpropane, 370 parts of hexahydrophthalic acid, 292 parts of adipic acid and 398 parts of isophthalic acid, followed by reacting at 220°C for 6 hours, adding 45 parts of trimellitic anhydride, reacting at 170°C for 30 minutes to obtain a polyester resin having a number average molecular weight of about 8000, acid value of 20 mg KOH/g and hydroxy value of 95 mg KOH/g.

(Note 2) Aliphatic hexafunctional blocked polyisocyanate compound prepared by blocking a trimer adduct of hexamethylene diisocyanate with methyl ethyl hetoxime.

(Note 3) The fine aluminum powder coated with a phosphate group-containing compound prepared by mixing 100 parts of a fine aluminum powder having a thickness of 0.03 to 0.1  $\mu\text{m}$  and a mean particle size of 5 to 6  $\mu\text{m}$  with 5 parts of the phosphate group-containing compound (Note 4) and 50 parts of cellosolve acetate, followed by leaving at rest at room temperature for 10 minutes to obtain a paste.

(Note 4) Phosphate group-containing compound:

A copolymer prepared from a monomer mixture of 30 parts of acid phosphoxyethyl methacrylate, 15 parts of 2-hydroxyethyl acrylate, 20 parts of methyl methacrylate, 5 parts of n-butyl methacrylate and 30 parts of 2-ethylhexyl methacrylate, and having a number average molecular weight of 13000 and hydroxy value of 72 mg KOH/g and acid value of 126 mg KOH/g.

(Note 5) Titanium oxide white pigment:

Titanium JR701 (trade name, marketed by Tayca

Corporation., Ltd., mean particle size of 0.3 to 0.6  $\mu\text{m}$ ).

#### Preparation Experiment 5

Preparation of water based intercoat coating composition No. 2 (corresponding to water based intercoat coating composition (B<sub>1</sub>-iii)):

Preparation Experiment 4 was duplicated except that the fine aluminum powder (Note 3) and the titanium oxide white pigment (Note 5) added in the water based intercoat coating composition No. 1 were not added to obtain the water based intercoat coating composition No. 2.

#### Experiment 1 (corresponding to Example 1 in the present application) and Comparative Experiments 1-3

A zinc phosphate treated dull finish steel plate was dipped into electrodeposition coating bathes containing the cationic electrodeposition coating compositions No. 1 to No. 3 as cathodes respectively, followed by carrying out an electrodeposition coating for 3 minutes at 30°C and 200 V so as to be a coating film thickness of 25  $\mu\text{m}$ , drying at 100°C for 10 minutes, coating thereonto water based intercoat coating compositions No. 1 to No. 2 by airless spray coating so as to be a coating film thickness of 15  $\mu\text{m}$  as shown in Table I, and heating and crosslink-curing both coating films simultaneously.

The resulting multi-layer coating films were subjected to coating film performance tests. Test results are shown in Table I.

Table I

	Experiment	Comparative Experiments		
	1	1	2	3
Cationic electrodeposition coating composition	No. 1	No. 2	No. 3	No. 1
Water based intercoat coating composition	No. 1	No. 1	No. 1	No. 2
Gloss	88	79	80	85
Smoothness	3	2	2	3
Anti-chipping properties	3	3	3	3
Weather resistance	3	3	3	1
Interlayer adhesion properties	100	100	100	0

Test Method

Gloss: 60° specular reflectance

Smoothness: Coating film appearance was visually evaluated as follows. 3: Good in smoothness, 2: slightly poor in smoothness, 1: seriously poor in smoothness.

Anti-chipping properties:

By use of a Q-G-R Gravelometer (trade name, marketed by Q Panel Co., Ltd.), 100g of gravels having a diameter of 15 to 20 mm was air sprayed onto the surface of a coating film at a spray angle of 90°, under an air pressure of about 4 kg/cm<sup>2</sup> at -20°C, followed by visually evaluating conditions of the surface of the coating film as follows. 3: slight impact mars in the surface of an intercoat coating film, but no separation of an electrodeposition coating film; 2: considerable amount of impact mars on the surface of the intercoat coating film, but slight separation of the electrodeposition coating film; 1: many impact mars on the surface of the intercoat coating film, and considerable

separation of the electrodeposition coating film.

Weather resistance:

Irradiation of 1000 hours by use of a sunshine weatherometer was followed by visually evaluating conditions of the surface of the coating film as follows. 3: showing no changes and good weather resistance; 2: showing reduction in gloss, slight cracks, chalking or whitening, etc., and slightly poor weather resistance; 1: showing reduction in gloss, remarkable cracks, chalking or whitening, etc., and poor weather resistance.

Water resistance:

Dipping into hot water at 80°C for 24 hours was followed by washing with water, and drying to visually evaluate the surface of the coating film as follows. 3: nothing abnormal; 2: slight reduction in gloss; 1: whitening developed.

Interlayer adhesion properties:

The above dipping into hot water was followed by cutting by use of a cutter so as to reach a substrate and to form 100 squares of 1 mm x 1 mm, followed by adhering thereonto an adhesive cellophane tape, strongly separating the tape at 20°C, and examining a number of remaining squares.

Conclusion

Comparison of Experiment 1 with Comparative Experiments 1 and 2 shows that combinations of the cationic electrodeposition coating composition No. 2 containing bismuth lactate as cited in USP5702581 (Reference 3) or the

cationic electrodeposition coating composition No. 3 containing  $\text{Bi}_2\text{O}_3$  with the water based intercoat coating composition No. 1 show unsatisfactory properties in gloss and smoothness.

Further comparison of Experiment 1 with Comparative Experiment 3 shows that a combination of a cationic electrodeposition coating composition containing bismuth hydroxide with a water based intercoat coating composition containing a fine aluminum powder makes it possible to obtain a multi-layer coating film showing good properties in all of gloss, smoothness, anti-chipping properties, weather resistance and interlayer adhesion properties.

4. The undersigned Petitioner declares further that all statements made herein of his own knowledge are true and that all statements made in information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

5. Further, Deponent saith not.

Date: October 27, 2003

Syuichi Ikenoue

Signature: Syuichi Ikenoue.